REPORT No. 33.

SELF-LUMINOUS MATERIALS.

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A limited mimeographed edition of the following notes was issued August 10, 1918, as Luminous Material Circular No. RLC-6 of the National Bureau of Standards. The cumbrous title of that circular—"Notes and Recommendations regarding Specifications for the Illuminating of Articles by Means of Self-Luminous Materials Containing Radioactive Excitants"—indicates its purpose and scope.

The circular was prepared with special reference to the illumination of military and of naval instruments. In its preparation the numerous questions propounded to the Bureau during the preceding 12 months in regard to such illumination were kept constantly in mind, the greater portion of its contents bearing directly upon these questions.

1. *Basis of specifications.*—As the object of illuminating articles with self-luminous materials is to secure a sufficient brightness combined with a satisfactory life, it is most desirable that the specifications be based upon the brightness at delivery and upon the rate of decrease in the brightness rather than upon such a subsidiary consideration as the amount of radioactive material contained in the preparation. This becomes at once evident when it is recognized that the brightness of a given specimen of self-luminous material depends not only upon the amount of the radioactive constituent, but also upon the age of the material and upon the properties of its phosphorescent responsive base. It is emphasized by the fact that the brightness of the applied material varies greatly with the method of application.

2. *Self-luminous material*—(*a*) *Cause of luminescence.*—Self-luminous radioactive materials consist of mixtures of a responsive base, usually zinc sulphide, with small amounts of radioactive substances. Luminescence is caused and maintained by the bombardment of the responsive base by the alpha radiations accompanying the disintegration of the radioactive constituents of the material. Other things being the same, the brightness of the material increases as the intensity of the alpha radiation increases. The actual brightness depends upon the responsiveness of the base as well as upon the intensity of the bombardment.

(*b*) *Secular changes in alpha radiation.*—The decrease in alpha radiation due to radioactive disintegration in six months is 0.02 per cent for radium, 2 per cent for radium-D, 6 per cent for mesothorium-1, 16 per cent for radiothorium, and 60 per cent for polonium. These values assume practical equilibrium between the elements named and their complete series of radioactive derivatives. This assumption is fulfilled for polonium (it has no such derivatives), for radiothorium, for radium after a month if the preparation has the equilibrium amount of radium-D and of polonium, for radium-D from which no polonium has been removed for at least four years, and for mesothorium from which no radiothorium has been removed for at least 15 or 20 years.

If radiothorium has been removed from the mesothorium within the preceding 15 or 20 years, the secular decrease in the alpha radiation will be less than 6 per cent. To be exact, after a complete removal of radiothorium the alpha radiation of a mesothorium preparation is zero; it at once begins to increase, and continues to grow for between four and five years; it

then decreases at a rate that is at first very slow, and finally after 15 or 20 years becomes practically 6 per cent in six months. From the third year to them iddle of the fifth year the intensity increases by a total of 7.4 per cent and by the seventh year it has fallen about 2 per cent below what it was at the third year.

After a complete removal of polonium, the alpha radiation of a radium-D preparation is zero; it at once begins to increase, and continues to grow for a period of about two years. It then decreases at a rate that is at first slow, and finally, after the fourth year (counted from the time the polonium was removed), becomes practically 2 per cent in six months. From the eighteenth to the twenty-fourth month the intensity increases by a total of 2 per cent and by the thirty-sixth month it has fallen to approximately what it was at the beginning of the nineteenth month.

In the preparation of material containing radium there is a considerable escape of radium emanation,[1] there is also a loss of radium emanation during the application of the material to the article being illuminated. This reduces the intensity of the alpha radiation. As the radium emanation again accumulates the intensity increases; the recovery is half completed in 3.85 days and is practically complete by the end of a month.

If the radium salt when incorporated with the material contained no radium-D or polonium its alpha radiation at the end of a month will be about 20 per cent less intense than if it had contained the equilibrium amounts of these substances, and will slowly increase for over a hundred years. The rate of growth, beginning at a very small value, increases rapidly for the first few months, then more slowly until at the end of two years the rate of growth will be about 0.4 per cent in six months, and will then gradually decrease to zero in about 115 years, at which time the total alpha radiation will be about 16 per cent greater than it was at the end of the first month.

If material containing radium, mesothorium, or radiothorium is not hermetically sealed, any increase in temperature will cause some of the air contained in it to escape, carrying with it a portion of the radium, or thorium, emanation. This will reduce the intensity of the alpha radiation; on keeping the temperature constant there will be a recovery. For radium the the recovery will proceed as just described, being half completed in 3.85 days. For the thorium products the recovery is very rapid, being one-half completed in less than a minute. Not only is the recovery of the alpha radiation of the thorium products very rapid, but, on account of the extremely short life of thorium emanation, the initial reduction is less than for the radium products, most of the thorium emanation disintegrating before it can escape from the material.

A reduction of pressure produces an effect of the same kind as an elevation of temperature.

Exclusive of such subsidiary effects as those of temperature and pressure, the secular variation in the intensity of the alpha radiation of self-luminous materials may be briefly described as follows: If the radioactive constituent is *radium* with its equilibrium amounts of radium-D and polonium, the intensity will increase for a month, rapidly at first then more slowly; after a little over a month the intensity will decrease very slowly, about 0.02 per cent in six months. If initially there was no radium-D nor polonium, the intensity will continue to increase after the first month; the rate of increase slowly decreasing, but for many years being about 0.4 per cent in six months. If the radioactive constituent is *radium*-D, the intensity will continually increase until about two years have elapsed since the removal of polonium, then the intensity will decrease, the rate of decrease never exceeding about 2 per cent in six months and attaining this value only after four years have elapsed since the separation of the polonium. If the radioactive constituent is *mesothorium-1*, the intensity will continually increase until about 4.5 years have elapsed since the removal of the radiothorium, then the intensity will decrease, the rate of decrease never exceeding 6 per cent in six months and attaining this value only after 15 or 20 years have elapsed since the separation of the radiothorium.

If the radioactive constituent is either *radiothorium* or *polonium*, the intensity will continually decrease, the rate being 16 and 60 per cent, respectively, in six months. All these values assume that the radioactive materials mentioned are not accompanied by their radioactive predecessors.

(c) *Secular changes in responsive base.*—Under the action of light, air, humidity, alpha ray bombardment, etc., changes take place in the responsive base. These changes are accompanied by a change, usually a decrease, in responsiveness. The most striking of these changes is that due to the alpha bombardment. Under continued alpha radiation the responsiveness decreases, very rapidly at first, then more and more slowly as the responsiveness becomes less. Other things being the same, the more intense the radiation the more rapid the decrease in the responsiveness. Whether the responsiveness will ultimately become zero is an open question. The nature and extent of the changes just described are functions of the responsive base and vary with the latter; the base generally employed is zinc sulphide, and it is this that we have in mind throughout these notes.

(d) *Secular changes in brightness.*—The secular variation in the brightness of self-luminous materials is due to the two causes just considered, viz, (1) the variation in the intensity of the alpha radiation, and (2) the secular changes in the responsive base. For practical purposes the resultant effect may be regarded as the sum of the changes due to these two causes.

If the material contains radium, the initial increase in the intensity of the alpha radiation more than offsets the effect of the decrease in the responsiveness. As a result, the brightness increases rapidly for about three weeks, then decreases for about two weeks at a rate much greater than that which later prevails.

If the radioactive constituent is one of the other substances mentioned, the brightness shows no initial growth,[1] but decreases continually from the start; first rapidly, then more slowly.

In general we may say that the brightness of a self-luminous material undergoes marked changes during the first four or five weeks after the radioactive constituent is incorporated in it. After this period, the brightness decreases at a more moderate rate. Of material containing the same responsive base the brighter the material the more quickly does its brightness drop to 50 per cent of its former value. For this reason it is desirable to design the illuminated portions in such a manner that a material of low brightness can be used.

No exact figure can be given for the rate at which the brightness of applied material may be expected to decrease. Wide variations in the rate are found even among dials illuminated at the same time and with the same material. No explanation of this variability can now be given.

Dials of 5 microlamberts illuminated with radium material have been observed to decrease to 2.5 in six months; others have decreased to only 3 or 4. Similarly dials of 8 may decrease to 3 or to 6 in the same time, and those of 2 to 1.3 or to 1.9.

Dry material containing radium and sealed in glass tubes may be expected to decrease approximately as shown in the following table:

TABLE 1.—*Decrease in brightness of dry material in 180 days.*

Brightness.	Brightness after 180 days.	Decrease in 180 days.	Brightness.	Brightness after 180 days.	Decrease in 180 days.
Microlamberts.	*Microlamberts.*	*Per cent.*	*Microlamberts.*	*Microlamberts.*	*Per cent.*
30	16.1	46	10	7.8	22
25	14.5	42	8	6.5	19
20	12.7	36	5	4.4	12
15	10.5	30	2	1.9	5

(e) *Brightness reduced by adhesive.*—The brightness of material that has been mixed with an adhesive and painted upon an object is usually only 25 per cent, occasionally 40 per cent, of that of the dry material. We have already seen that dry material decreases in brightness at a lower rate than does painted material of the same brightness. It is, therefore, desirable that dry material be used wherever possible.

3. *Unit of brightness.*—By the brightness of a surface is meant the luminous intensity per unit area of the surface as projected on a plane perpendicular to the line of sight, it being assumed

[1] Mesothorium recently freed from radiothorium will give an initial increase in brightness; the rate and duration of this increase will depend upon the time that has elapsed since the separation of the radiothorium. It is very improbable that such fresh mesothorium will ever be used in the preparation of commercial luminous material.

that the linear dimensions of the portion of the surface considered is negligibly small as compared with the distance at which it is viewed. It is evident that the brightness as thus defined is independent of the area of the luminous surface.

As in measuring a length we choose for our unit a length that is exactly defined (foot, meter, etc.), so in measuring a brightness we choose as our unit the brightness of an exactly defined bright surface.

For the measurement of the brightness of self-luminous materials a suitable unit is the brightness of a perfectly diffusing and completely reflecting surface when illuminated by a source of unit candlepower placed at a distance of 10 meters. This unit of brightness is called a *microlambert* because it is the one-millionth part of the lambert; the latter is a well-known and accepted unit of brightness. The microlambert is equal to 0.000 93 "equivalent foot-candles" and to 0.000 000 318 candles per square centimeter (3.18 millicandles per square meter).

4. *Total light.*—It is necessary to distinguish between the brightness of a luminous surface and the total amount of light the eye receives from it. The former is determined by the luminous material, while the latter depends also upon the area of the luminous surface. When the luminous area is small as compared with its distance from the eye it is the total light that gives the impression of brilliance and determines the ease with which the luminous area can be seen.

5. *Brightness of markings.*—(*a*) *Position marks.*—Marks that have to be seen, but of which the shapes do not have to be distinguished, require only a low brightness if they are to be viewed against an extended black background. Lines 1 millimeter wide and 4 millimeters long upon a black metal surface in moonlight are not readily seen in all positions at a distance of 75 centimeters (30 inches) if they are no brighter than 2 microlamberts. If the surface and the observer's eyes are shaded from the moon then such lines are seen without difficulty.

(*b*) *Legible character.*—If the luminous markings (numerals, letters, etc.) are of such a character that their shapes have to be distinguished, the brightness must be greater than that required for the mere seeing of position marks. Complex characters require a higher brightness than simpler ones of the same size. Numerals 8 millimeters high with a line width of 1 millimeter can be readily read in the moonlight at a distance of 75 centimeters (30 inches) if they have a brightness of 5 microlamberts.

(*c*) *Relation of brightness to size.*—There are at least four distinct phenomena that limit the distance at which a luminous character can be read:

First. The resolving power of the eye. Unless the angle the character subtends at the eye exceeds a certain value, the shape can not be recognized, however bright the character may be. This limiting angle varies greatly from eye to eye.

Second. The sensitivity of the eye. This also varies over a wide range and depends upon both the past and the present history of the eye. An eye that has recently been exposed to light is far less sensitive than if it had been kept in total darkness for some time; also, if the eye is illuminated by an extraneous source of light its sensitivity is different from what it is when the eye is illuminated by the luminous character alone.

Third. Contrast. It is evident that the character can not be distinguished if there is no contrast between it and the region bounding it. The percentage contrast in brightness that can be just distinguished increases as the brightness is reduced. For the range of brightness covered by articles illuminated by self-luminous materials this limiting contrast varies approximately from 5 to 20 per cent. That is, very faint markings will not be seen unless they are at least 20 per cent brighter than the surrounding surface. In most practical cases the lines are illuminated by the same light as the surface on which they are painted; their effective brightness is therefore enhanced. This, however, may not offset the ill effect of the light from the main surface of the article. For example, a white dial illuminated directly by the full moon might have a brightness of 20 microlamberts. If the marks upon it had a brightness of 1 microlambert and a coefficient of reflection as great as that of the dial their effective brightness would be but 21 microlamberts. This is but 5 per cent greater than the brightness of the main surface of the dial, and the marks would be almost if not entirely indistinguishable.

Fourth. A fourth limit is set by the total amount of light entering the eye from the character. Assuming that the conditions are such that this fourth limit is the only one operative, then for the same surface brightness a given character can be distinguished twice as far if all its dimensions are doubled; if its size is kept the same its brightness must be multiplied by 4 if it is to be distinguished at twice its former distance. Expressed otherwise, this fourth limit states that the distance at which a given character can be distinguished is proportional to its linear dimensions multiplied by the square root of its brightness. As the determining factors are the brightness and the angle which the object subtends at the eye, the effect of increased size can be secured by the use of a magnifyng lens.

One result of this fourth limiting condition is that neighboring luminous marks may appear to blur together although they are much farther apart than is necessary to enable one to see them as distinct under usual conditions of illumination. For ready seeing at low brightness luminous characters should be not only large, but open. There should be as few small inclosed unilluminated areas as possible; for example, unless the 3, the top of the 6, and the bottom of the 9 are well opened, there is danger of mistaking these numerals for 8's.

(*d*) *Guides to required brightness.*—The brightness needed in any particular case should be determined by actual experiment. Objects of the required form should be illuminated to various brightnesses and examined under conditions of illumination that are as nearly as may be identical with those met with in service. Frequently a stencil illuminated from the rear serves admirably. When the object is illuminated with luminous material it is desirable that it be made too bright and then dimmed down by the use of absorbing screens.

In such tests at least two values should be determined: (1) The lowest usable brightness; (2) The brightness that is considered the most desirable. If there is a maximum brightness that must not be exceeded this should be determined also.

The various values of the brightness determined by these tests should be measured in terms of an established unit, preferably the microlambert.

The following conclusions, drawn from observations made by various observers under a sky lighted by a full moon, may serve as a rough guide to the order of brightness that probably will be required. These conclusions apply to luminous lines on a black surface. They are but rough approximations, different observers and different conditions of general illumination leading to different conclusions.

Division marks of one microlambert are too faint unless the lines are very broad, those of 1.5 microlamberts are usable, but 2 microlamberts or over is to be preferred. If the lines are 4 millimeters long and 1 millimeter wide, and have a brightness of only 2 microlamberts there are some positions in which they can not be readily seen at 75 centimeters in moonlight.

Numerals 7 millimeters high with a line width of 0.6 millimeter and a brightness of 2 microlamberts can be read at a distance of 30 centimeters (1 foot), but a brightness of 4 is much better. Numerals 8 millimeters high with lines 1 millimeter wide are readily read at 75 centimeters ($2\frac{1}{2}$ feet) if they have a brightness of 5.

A circular strip 6.4 millimeters ($\frac{1}{4}$ inch) wide with outside diameter of 38 millimeters ($1\frac{1}{2}$ inches) is visible at 9 meters (30 feet) if it has a brightness of 6 microlamberts.

6. *Summary.*—In designing an article to be illuminated, and in drawing specifications for the illumination of it, the following points should be considered:

(*a*) The characters should be large and of simple design; usually they should be as large as is consistent with the other requirements. Published results indicate that when the ratio of height of character to width of line is equal to 8 the best condition for legibility of such letters as N, S, E, and W has been secured.

(*b*) The spacing should be open. It is undesirable to reduce the distance between adjacent luminous areas to less than 2 millimeters.

(*c*) The number of luminous markings should be reduced to a minimum.

(*d*) Pointers can advantageously be luminous from pivot to tip, but the tail should never be illuminated. In many cases it is unnecessary for the luminous material to cover the entire width of the pointer.

(*e*) Unilluminated areas that can be seen by the observer should be dull black.

(*f*) The brightness required should be determined in each case by an actual trial under service conditions of illumination and distance.

(*g*) The nature of the radioactive constituent of the material will be determined by the life requirements. If the life is to exceed six months, polonium unaccompanied by an equilibrium amount of radium-D should not be used; and only under special conditions would radiothorium be satisfactory; there would be but little choice between mesothorium-1 and radium. If a short life of high brightness is desired, radiothorium, radium-D, and, in certain cases, polonium, might give satisfaction; the life of very bright material is in any case short on account of the rapid loss in the responsiveness of the base.

(*h*) The secular decrease in the brightness of the material is very perceptible and must be allowed for.

(*i*) The specifications should be so worded as to—

Prevent the use of undesirable radioactive constituents. (See caution, p. 9.)

Specify the brightness of each part on delivery.

Specify that the brightness shall not decrease by more than a specified amount in a specified time. When definite limits of this kind can not be given, it should be specified that the brightness shall require the longest period practicable to decrease to one-half its value on delivery.

Require that the material, especially if it contains radium, shall have been completely prepared for several, preferably at least five, weeks before delivery.

Provide for inspection on delivery, and for the selection by the inspector of specimens for submittal to a life test.[1]

Require that the manner of application shall be suitable for the purpose.

State clearly the size, design, and position of each luminous mark.

Require the bidder and the contractor to state in every case (1) the trade name and grade of the material used; (2) the manufacturer of the material, and (3) the firm that applies the material.

7. *Appendix.*—(*a*) *Caution.*—Since the only function of the radioactive constituent of a self-luminous material is to furnish an alpha ray bombardment for exciting the luminescence of the responsive base, the effective measure of any constituent is not its weight but the intensity of the alpha radiation furnished by it. The intensity of the radiation from a short-lived material is much greater than that from an equal weight of a long-lived one. Illustrations of this difference are given in the following table.

TABLE 2.—*Comparison of the rates of emission of alpha particles.*

[From certain radio-elements as compared with their rate of emission from radium with its derivatives to and including radium-C.]

Radio-element.	Radium.
	Milligram.
1 true milligram mesothorium-1, with derivatives	= 450
1 true milligram radio-thorium, with derivatives	=1,050
1 true milligram polonium (radium-F)	=1,200

On account of this fact it is most necessary to exercise extreme care in the wording of those portions of specifications and other discussions of luminous materials that deal with the relative amounts of different radio-elements contained in the material.

For example, the statement that "75 per cent of the radioactive constituents shall be radium" is most ambiguous. In order to be of value in determining the properties of the luminous material the restriction should state explicitly that "75 per cent of the alpha radiation of the material shall be due to radium." Otherwise the restriction may be interpreted as meaning 75 per cent by weight. It is evident from the table that the presence of 25 per cent by weight of polonium will give the material practically the same characteristics as if activated entirely by polonium.

[1] The Bureau of Standards is prepared to conduct such life tests for the Government, and, on request, will consider the undertaking of similar tests for the public.

(*b*) *Alpha radiation—Time curves.*—Figures 1, 2, 3, and 4 show the secular variation in the alpha radiation from material containing mesothorium–1, radium–D, radiothorium, and polonium, respectively.

The constants used in the computation of these curves are those given by Rutherford in his "Radioactive substances and their radiations."

(*c*) *Brightness—Time curves.*—Figures 5, 6, and 7 represent the variation in brightness of four tubes of dry radium luminous material studied at this Bureau. The four specimens are believed to differ only in the amount of radium contained in them. The radium was added to the zinc sulphide only a few days before the first observation, somewhere about the thirtieth day from the origin of time arbitrarily chosen for these curves. Throughout the period covered by these curves the tubes were protected from external light.

(*d.*) *Form of specifications for the illumination of articles by means of self-luminous materials containing radioactive excitants.*

GENERAL.

1. These specifications cover the requirements of ------ for the illumination of ------

2. When luminous ------ are ordered, the luminous material used must conform to these specifications. The markings must conform to those indicated in the specifications for the individual articles.

3. The manufacturer of the luminous articles shall furnish the following with his proposal:

(*a*) The trade name and grade, and the name of the manufacturer of the luminous material he intends to use.

(*b*) The name of the company that will apply the material.

(*c*) Samples of the proposed illumination. These samples will be subjected to examination and tests, and may not be returned to the bidder.

4. The manufacturer shall notify ------ of any change in the luminous material from that accepted.

5. Whenever the word "brightness" occurs in these specifications it shall be understood to mean the surface brightness of the part after it has been continuously protected from all external light for the immediately preceding period of eight hours.

MATERIAL.

6. The luminous material shall consist of a suitable compound containing radium or mesothorium-1, or both. No radioactive material of a shorter average life than mesthorium-1 (7.9 years) shall be present unless accompanied by at least the equilibrium amount of its immediate predecessor in the same radioactive series. (See p. 9.)

7. No radioactive substance shall have been added to the luminous material during the five weeks preceding the inspection for the acceptance of the illuminated articles.

8. The quality and the composition of the luminous material shall be such that during the ------ months following the acceptance inspection the brightness of the illuminated article shall not decrease under service conditions by more than ------ per cent of its value at the time of such inspection. (Or, the quality and the composition of the luminous material shall be such as under service conditions to require the longest period practicable for the brightness of the illuminated article to decrease to one-half its value at the time of the acceptance inspection.)

BRIGHTNESS.

9. At the time of the inspection for acceptance the brightness of each part shall be as stated in the following schedule:

Schedule of brightness.

Article and part.	Brightness.	Remarks.
	Microlamberts.	
....	....	....
....	....	....
....	....	....

APPLICATION.

10. The luminous material shall be applied in such a manner that it will not become loosened or detached under normal service conditions.

11. Upon luminous parts that are to be immersed in a liquid, the luminous material must be so applied that it will not deteriorate from exposure to the liquid.

12 The surfaces to be covered with the luminous material will preferably be recessed or depressed, the material being placed in the cavities thus formed.

13. The surfaces to be covered with the luminous material will preferably be white, and shall be so finished as to prevent deleterious chemical action upon the luminous material.

14. Whenever practical, dry material (i. e., material unmixed with adhesive) will preferably be used, the material being kept in place by means of a transparent covering or container.

(The contract for the illumination of the articles will include in addition to the foregoing specifications the following provisions.)

INSPECTION.

15. All luminous parts shall be subject to inspection at .... by an agent designated by ....

16. The inspection of the luminous parts will include workmanship, brightness, uniformity, and such other points as may be deemed desirable.

TESTING.

17. The inspector shall select for further tests such samples as may be deemed desirable. This testing will cover the brightness, life, and durability of the illumination. These samples shall be sent to the place designated for such test, the expense of transportation being borne by ....

PENALTY.

18. (No recommendation.)

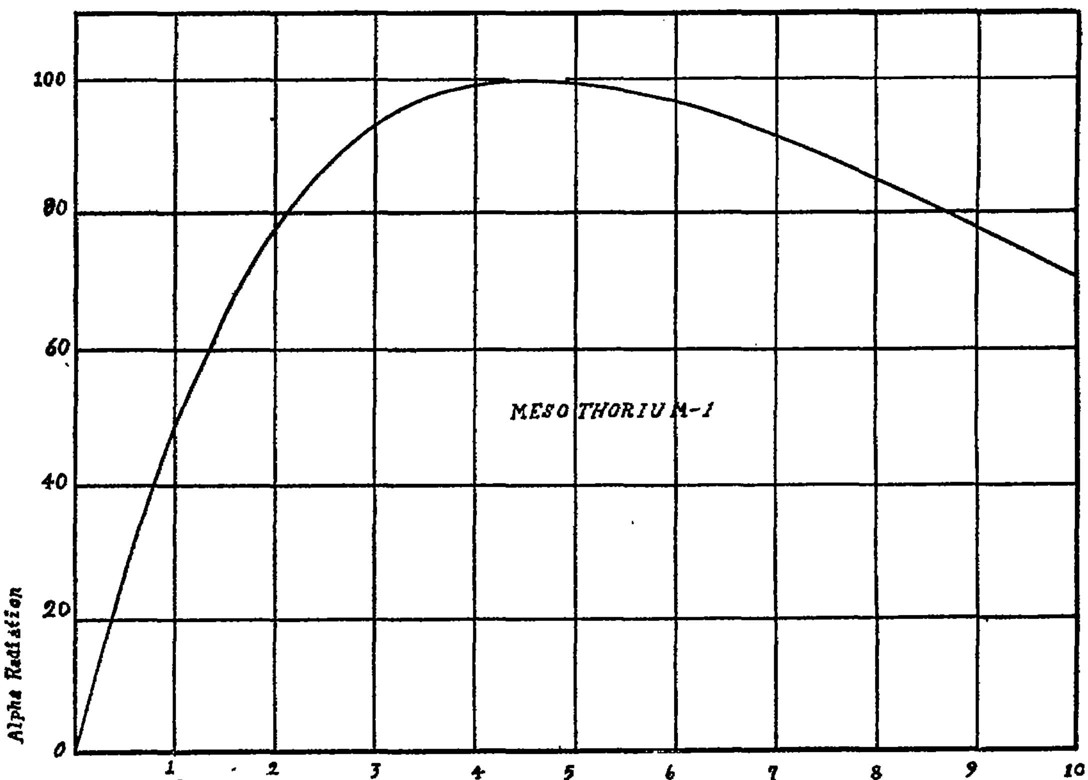

FIGURE 1.—Curve showing the secular variation in the alpha radiation from mesothorium-1 material. The time is counted from the beginning of the growth of radiothorium. The maximum intensity of the alpha radiation is arbitrarily chosen as 100.

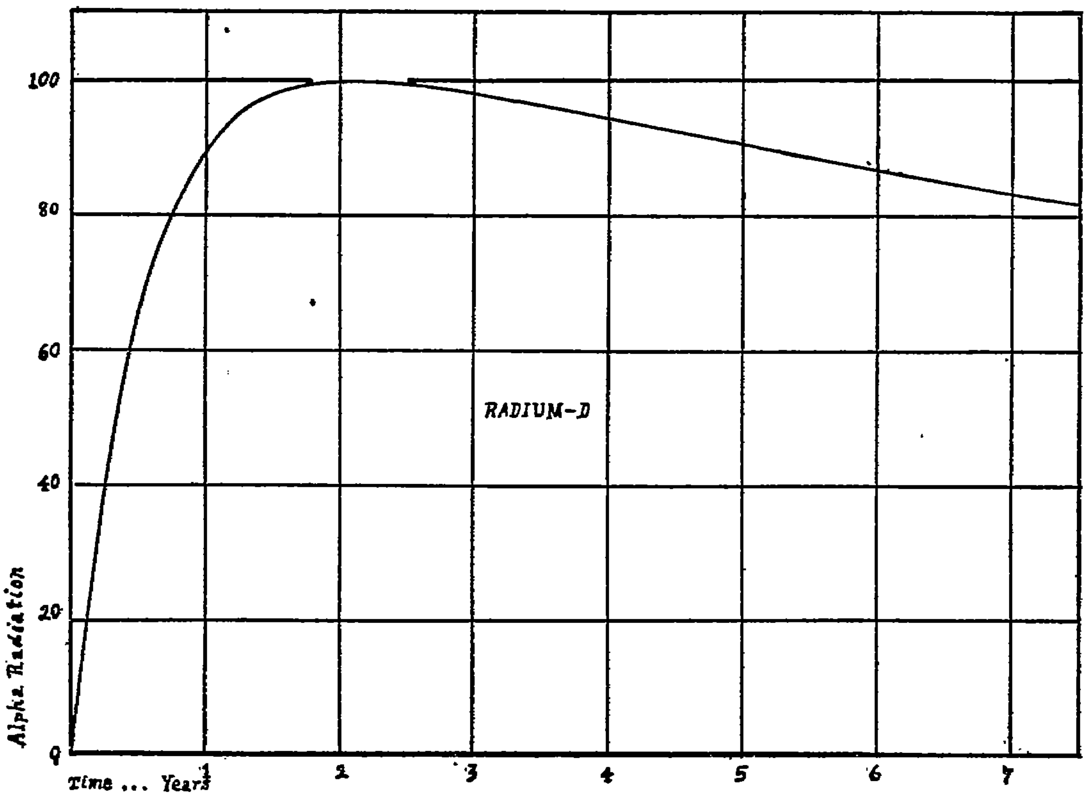

FIGURE 2.—Curve showing the secular variation in the alpha radiation from radium-D. The time is counted from the beginning of the growth of the polonium, and the maximum ordinate is chosen as 100.

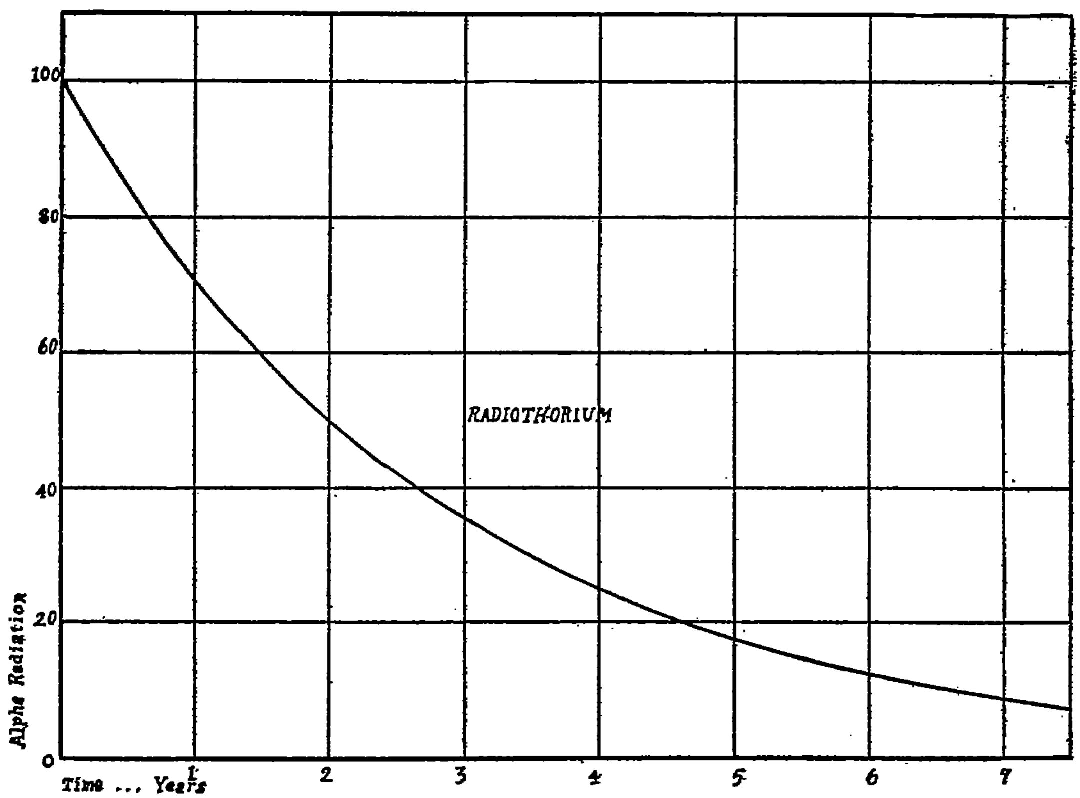

FIGURE 3.—Curve showing the secular variation in the alpha radiation from radiothorium.

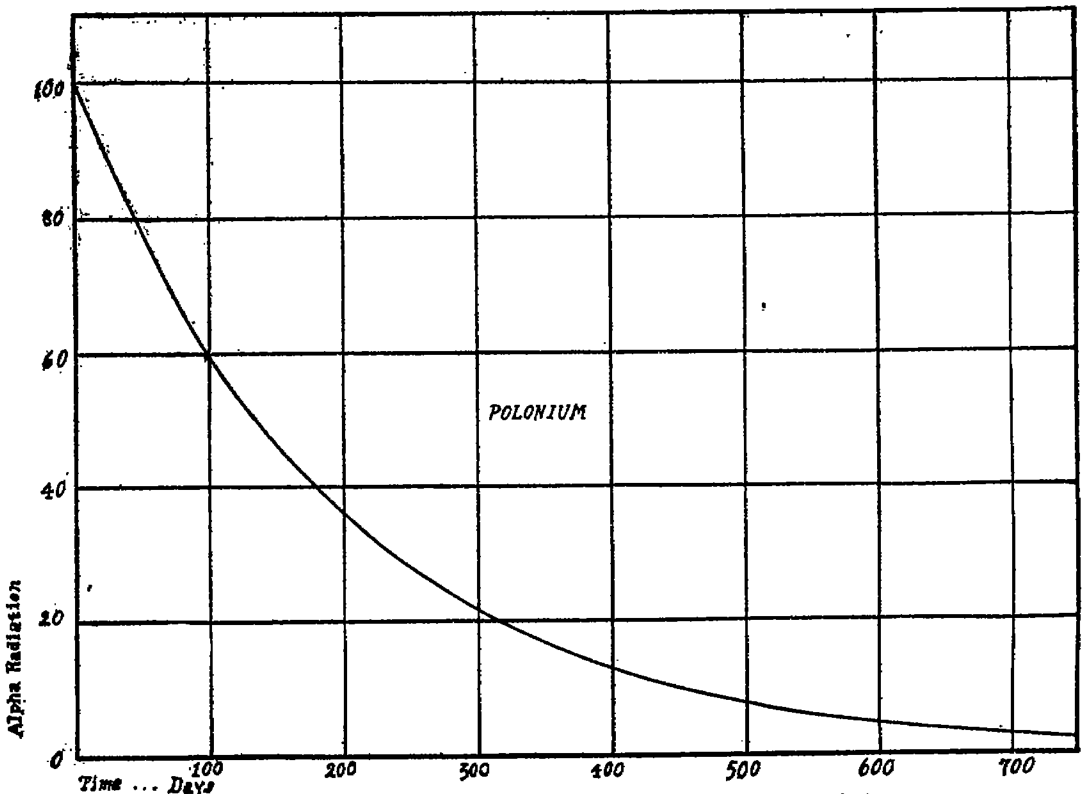

FIGURE 4.—Curve showing the secular variation in the alpha radiation from polonium.

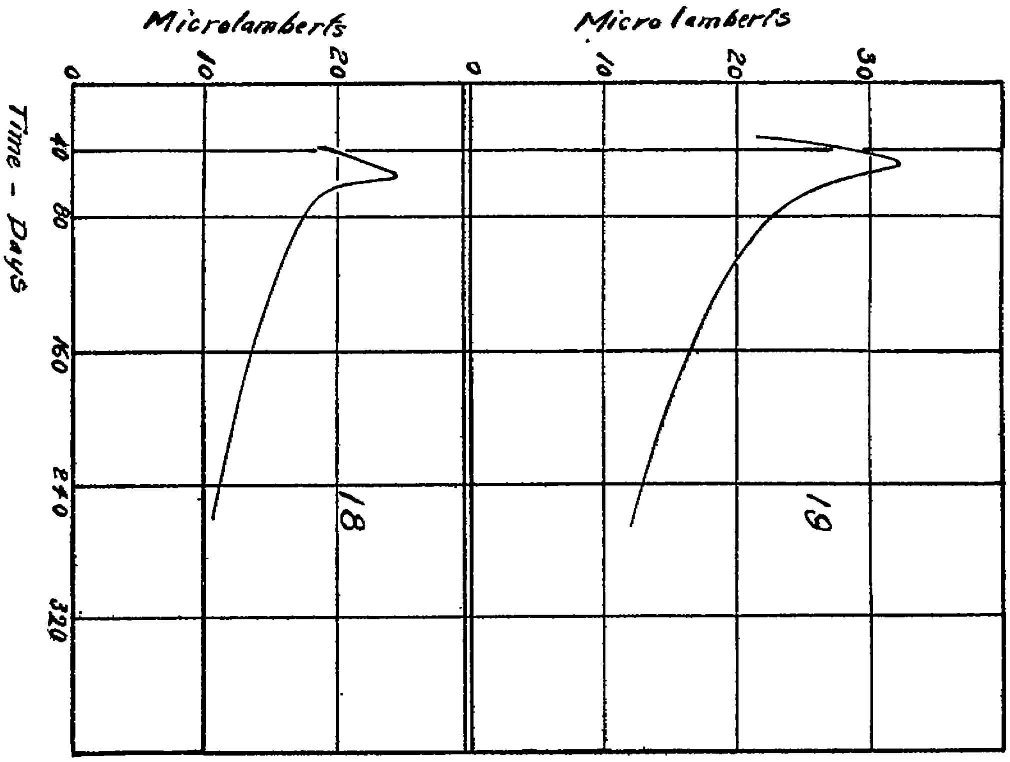

FIGURE 5.—Curves showing the variation in the brightness of two tubes of dry radium luminous material. The material represented by curve 18 contained 0.10 mg. of radium (element) per gram of material, that represented by curve 19 contained 0.15 mg. of radium per gram.

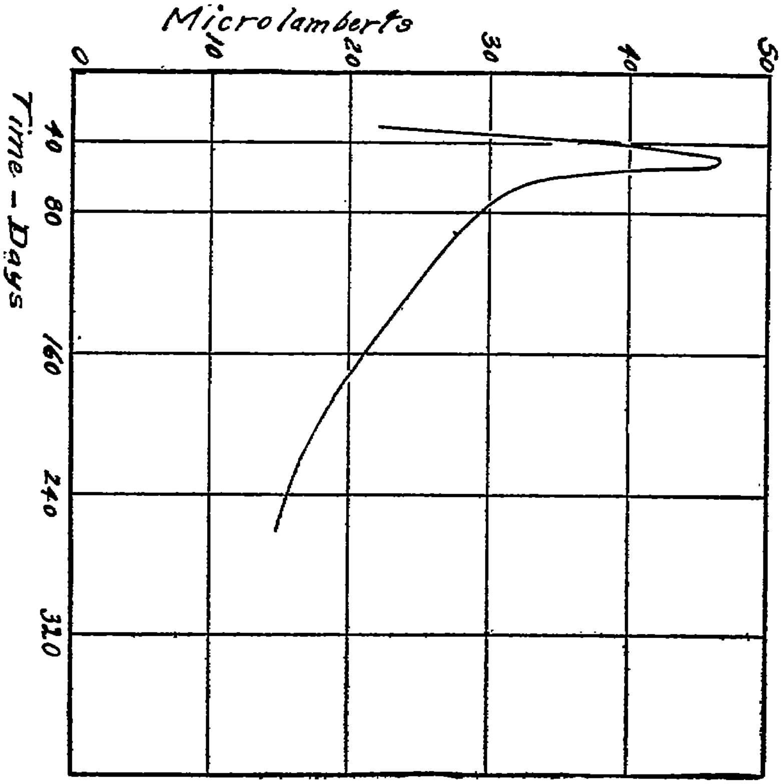

FIGURE 6.—Curve showing the variation in the brightness of a tube of dry radium luminous material containing 0.22 mg. of radium per gram.

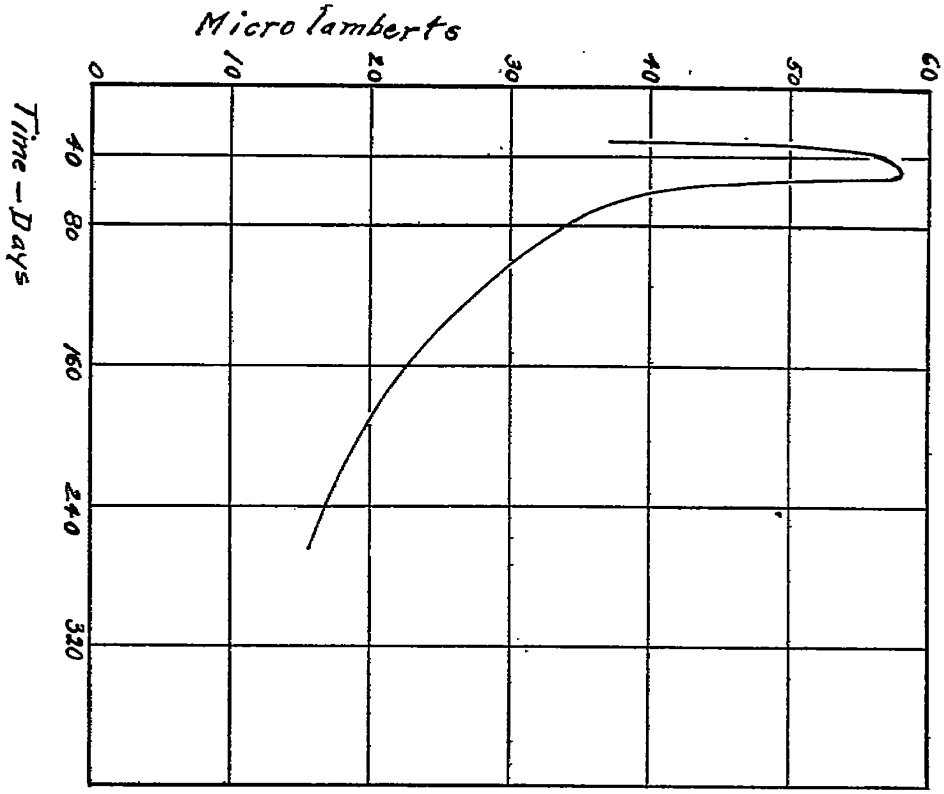

FIGURE 7.—Curve showing the variation in the brightness of a tube of dry radium luminous material containing 0.30 mg. of radium per gram.